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ABSTRACT

Utilizing a novel method with the resonance frequency of a LC circuit, we measured the superconducting anisotropy of single crystals of an Fe-based superconductor FeSe with applied magnetic field up to 16 T. We found that the temperature dependence of the upper critical field $H_{c2}(T)$ of FeSe coincides with the Werthamer-Helfand-Hohenberg (WHH) model when taking the Maki parameter α into consideration, suggesting an important role played by spin-paramagnetic effect in suppressing the superconductivity. When temperature $T \to 0$, the values of $H_{c2,\parallel c}(0)$ and $H_{c2,\parallel ab}(0)$ derived from the WHH fitting are close to and fall within the range of the Pauli limit, for field H_0 applied parallel to the c-axis and to the ab-plane, respectively. As compared with other typical iron-based high-T_c superconductors, lower values of $H_{c2}(0)$ and higher superconducting anisotropy $\Gamma(0)$ were observed in FeSe.

I. INTRODUCTION

Superconductivity in Fe-based superconductors has been widely studied since their first discovery in 2008 due to its potential application and the needs of its understanding in physics.[1] The family of Fe-based superconductors can be mainly categorized into "1111-type" RFeAs(O,F) (R = rare earth),[1–4] "122-type" material (e.g., $BaFe_2As_2$),[5,6] "111-type" LiFeAs,[7,8] and "11-type" iron chalcogenides[9,10] according to their crystal structure. In fact, these compounds have similar features in structure, such as a square-planar lattice of Fe with a tetrahedral coordination. Among them, iron selenide, FeSe, has the simplest structure and has similar Fermi surface topology to others.[11] Therefore, FeSe has attracted great attention as a model system to study the mechanism of high-T_c superconductivity in Fe-based superconductors.[12–14]

The upper critical field H_{c2} is one of the fundamental parameters for superconductivity, which can provide valuable information such as coherence length, penetration depth, and pair-breaking mechanism. Thus the investigation of these parameters is crucial for the understanding of high-T_c superconductivity and its potential applications. Traditionally, H_{c2} and T_c are mainly measured by techniques of electrical resistivity, magnetic susceptibility and specific heat.[9,15–18] However, each experimental technique has its own advantage and disadvantage, and they show differences in results in various cases.[17,19–22] Thus, novel measurement techniques are highly valuable. One of the good candidates is the recently developed resonance frequency technique, which utilizes the direct inductance change in a resonant LC circuit induced by the bulk magnetic susceptibility and/or electrical resistivity change of the sample during the appearance of a superconducting phase transition.[23–25] This novel method can sensitively track the effect of the superconducting vortex phases on the shielding of the radio frequency (RF) magnetic field in a sample coil and was used to determine H_{c2} and T_c.[25] Thus it is very different from other measurement techniques since what is directly measured is the resonant frequency, not the sample electrical resistivity or magnetic susceptibility. One of the advantages is that it has a good clear background in the sample normal state when an applied magnetic field is changed, and usually the resonant frequency increases rather sharply once the sample enters a superconducting vortex state.[25]

Therefore, to systematically study the superconducting parameters of the model iron-based superconductor FeSe with the help of this novel method is important. On the other hand, previous research showed that two-band features dominate the pair breaking with the influence of a spin-paramagnetic effect in FeSe single crystals at $H_0 \parallel (101)$ and $\perp(101)$.[20] However, it's not clear whether this effect dominates at other directions, such as at $H_0 \parallel c$ and $\parallel$ *a&b*, even though there were measurements of H_{c2} and T_c on FeSe by other experimental techniques.[26,27]

In this paper, we reported the measurements of the superconducting anisotropy involving temperature-dependent upper critical field $H_{c2}(T)$ of FeSe single crystals by a resonance frequency technique, which uses a capacitance tuning LC circuit newly designed by ourselves. We showed that the experimental data for both field directions at $H_0 \parallel c$ and $H_0 \parallel$ *a&b* coincide with the Werthamer-Helfand-Hohenberg (WHH) model[28] when we consider the Maki parameter α, suggesting an important role of spin-paramagnetic effect in the pair-breaking mechanism in the superconductivity of FeSe. The zero-temperature upper critical fields $H_{c2,\parallel c}(0)$ and $H_{c2,\parallel ab}(0)$ were derived to be 12 T and 29 T from the WHH fitting,[28] for $H_0 \parallel c$ and $H_0 \parallel$ *a&b*, respectively, which give an anisotropy parameter $\Gamma(0) \sim 2.4$. According to Ginzburg-Landau theory and the anisotropy relation,[29] the coherence length and penetration depth were also analyzed. Furthermore, lower upper critical field and somewhat higher superconducting anisotropy at zero temperature were observed in FeSe in comparison with those of other typical iron-based superconductors.

II. EXPERIMENT

Single crystals of FeSe were grown in evacuated quartz ampoules using the $AlCl_3$/KCl flux technique with a temperature gradient of 5 °C/cm along the ampoule length. The temperatures of the hot and cold ends used were 427 °C and 380 °C, respectively.[30] The high phase purity of FeSe single crystals was verified by x-ray diffraction. The c-axis was found to be perpendicular to the ab-plane with a tetragonal crystal structure at room temperature. A typical plate-like single crystal FeSe sample with lateral dimensions up to $3.3\times2.7\times0.1$ mm^3 was used for our measurements [Fig. 1(a)].

The inductance L_C was introduced by a coil which was made from 50 μm silver wire wound with ~ 18 turns [Fig. 1(b)]. A FeSe single crystal was put inside the coil that was attached to a rotatable sample holder on a goniometer, with the sample rotation axis to be located in the lattice *a&b*-plane and perpendicular to H_0.

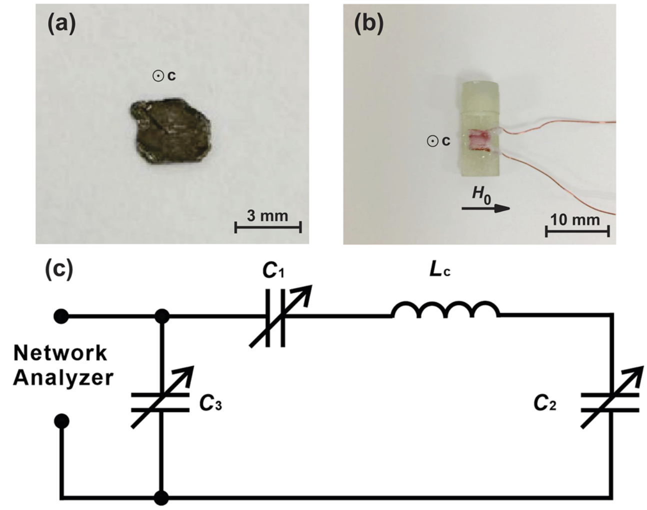

FIG. 1. Pictures of the measured FeSe single crystal sample (a) and the coil with the sample inside (b). (c) Sketch of the LC circuit used for the superconducting anisotropy measurements on FeSe.

The sketch of the capacitance tuning resonant LC circuit used for our superconducting anisotropy measurements is shown in Fig. 1(c). Besides the coil (L_C), the components of the circuit are two series capacitors (C_1, C_2) for tuning and one parallel capacitor (C_3) for matching the resonant frequency f_R, which are different from what we used earlier.[25] A commercial network analyzer was used to check the quality factor of this resonant circuit (up to 60) and to measure the circuit resonance frequency f_R. We set $f_R \sim 120$ MHz at room temperature, and measured the temperature dependence of f_R down to 1.8 K in different fields ranging from 0 to 16 T at both field alignments $H_0 \parallel c$ and $H_0 \parallel$ *a&b*.

III. RESULTS AND DISCUSSION

Figure 2 shows the measured resonance frequency f_R vs temperature T for FeSe in four typical magnetic fields at $H_0 \parallel c$. We can see that the f_R keeps constant when decreasing T until some specific temperature below 10 K is reached. Then f_R continues to increase upon cooling. Here we ruled out the possibility of the change of f_R from the coil itself since we repeated the same measurement without the FeSe sample being inside the coil, where no change of f_R was found.

Previous work has proved that an increase of f_R is originated from a change of the sample inductance as $f_R \sim 1/\sqrt{LC}$,[23–25] where L and C are the LC circuit inductance and capacitance, respectively. By neglecting the surface resistance of the sample, a simplified expression of L is: $L = L_C[1 + 4\pi\chi(T)]$, where L_C is the sample coil inductance itself and $\chi(T)$ is the temperature dependent sample magnetic susceptibility.[24,31,32] In other words, the increase of f_R represents the superconducting transition associated with a change in the magnetic susceptibility [a decrease in $\chi(T)$, for example, $\chi(T)$ can be negative] of the sample in the superconducting vortex state.[24]

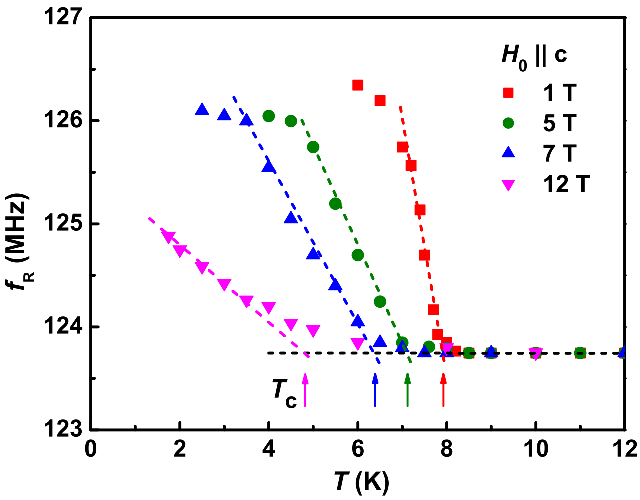

FIG. 2. The measured resonance frequency f_R versus T for FeSe in various applied magnetic fields at $H_0 \parallel c$. The dashed lines are used to determine T_c.

The data show that f_R increases with T at the superconducting transition. Thus T_c at different fields can be determined by the intersection of this linear fit (dashed lines and arrows in Fig. 2). We note that f_R starts to saturate upon cooling down to low enough temperatures as seen by the data at 1 T (not shown for all fields here).[25]

Similar measurements and analysis were also performed for the other field direction $H_0 \parallel a\&b$ (Fig. 3). The increase rate of f_R below T_c is similar to that at $H_0 \parallel c$, however, the value of T_c is higher and has less variation with the field. These differences indicate an existence of superconducting anisotropy in FeSe.

Furthermore, we systematically measured T_c at other values of magnetic fields, for both $H_0 \parallel c$ and $H_0 \parallel a\&b$ directions. Thus from

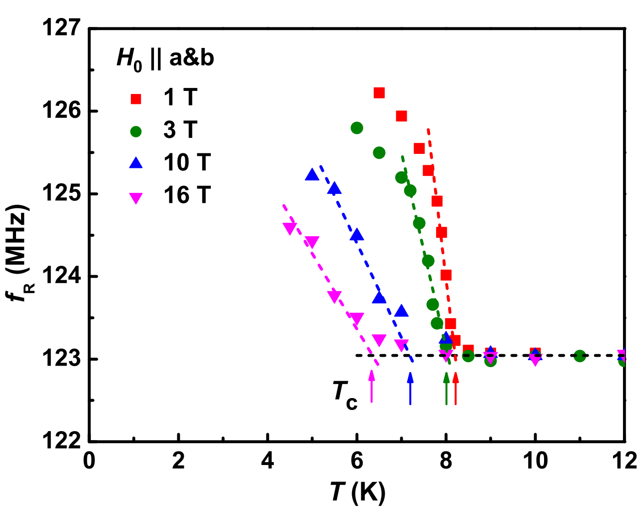

FIG. 3. The measured resonance frequency f_R versus T for FeSe in various applied magnetic fields at $H_0 \parallel a\&b$. The dashed lines are used to determine T_c.

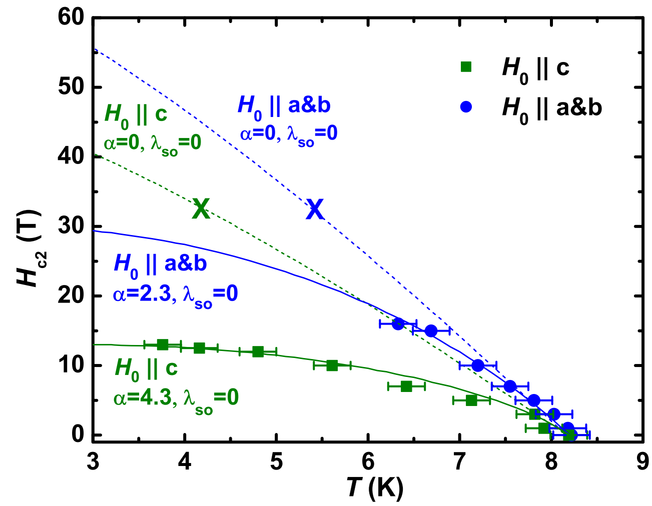

FIG. 4. The upper critical field H_{c2} versus T for FeSe with $H_0 \parallel c$ (green rectangular dots) and $H_0 \parallel a\&b$ (blue circular dots). The solid lines are the fits with spin-paramagnetic effect, which give the best fit, while the dashed lines (with cross signs) are the fits to the WHH model with $\alpha = 0$, $\lambda = 0$ (where the spin effect is ignored). (Note: our measurements can not go higher than 16 T due to the field limit of our magnet).

these, we obtained the phase diagram of the upper critical field H_{c2} vs T for FeSe, as shown in Fig. 4. Note, the values of H_{c2} correspond to the values of H_0 with $T = T_c$ as indicated by the arrows in Figs. 2 and 3. Noticeably, near T_c (H_0=0), the values of H_{c2} show linear increase with decreasing temperature, similar to previous work with other measurement methods.[15,17,18,20] Moreover, there is a saturation trend at lower temperatures, especially for $H_0 \parallel c$. An apparent difference between the values of H_{c2} at low temperatures for the $H_0 \parallel c$ and $H_0 \parallel a\&b$ directions is observed, indicating a rather high anisotropy.

In order to understand the superconducting anisotropy, we fitted the data with the WHH model,[28] which is usually used to describe the behavior of H_{c2} in typical two-dimensional type-II superconductors. According to this model, the upper critical field H_{c2}(T) can be calculated from the following equation in terms of digamma functions:[28,33]

$$\ln\frac{1}{t} = \left(\frac{1}{2} + \frac{i\lambda_{so}}{4\gamma}\right)\psi\left(\frac{1}{2} + \frac{\bar{h} + \lambda_{so}/2 + i\gamma}{2t}\right) + \left(\frac{1}{2} - \frac{i\lambda_{so}}{4\gamma}\right)\psi\left(\frac{1}{2} + \frac{\bar{h} + \lambda_{so}/2 - i\gamma}{2t}\right) - \psi\left(\frac{1}{2}\right), \quad (1)$$

where $t = T/T_c$, $\gamma \equiv [(\alpha\bar{h})^2 - (\lambda_{so}/2)^2]^{1/2}$ and $\bar{h} = 4H_{c2}/[\pi^2(-dH_{c2}/dt)_{t=1}]$. Here α and λ_{so} are the fitting parameters which correspond to spin-paramagnetic effect (also called Maki parameter[34]) and spin-orbit interactions, respectively. In the absence of both spin effects ($\alpha = 0$, and $\lambda_{so} = 0$), the equation can be simplified as[33]

$$\ln\frac{1}{t} = \psi\left(\frac{1}{2} + \frac{\bar{h}}{2t}\right) - \psi\left(\frac{1}{2}\right), \quad (2)$$

TABLE I. A comparison of T_c, upper critical field, anisotropy and coherence length at zero temperature of our FeSe sample with other typical Fe-based superconductors.

Materials	T_c (K)	$H_{c2,\parallel ab}(0)$ (T)	$H_{c2,\parallel c}(0)$ (T)	$\Gamma(0)$	$\xi_{ab}(0)$ (nm)	$\xi_c(0)$ (nm)
FeSe	8.2	29	12	2.4	5.2	2.2
$Fe_{1.1}Te_{0.6}Se_{0.4}$[33,35]	14	47	47	1	2.7	2.7
$K_{0.8}Fe_{1.76}Se_2$[36]	32	60 ~ 100	60	1 ~ 1.7	2.3	1.4 ~ 2.3
LiFeAs[37–39]	18	24	15	1.6	4.8	1.7
$(Ba,K)Fe_2As_2$ (10 K)[40]	28	57	55	1	2.2	2.2
KFe_2As_2[41]	3.6	7	1.4	5	15.2	3
$Ca_{0.83}La_{0.17}FeSe_2$[42]	41	85	47	1.8	2.7	1.5
FeS[43,44]	4.5	2.4	0.42	5.7	28	5
$Fe_{1.06}Te_{0.88}S_{0.14}$[45]	7	56	44	1.3	2.4	2.7

and it results in an orbital-limited value of upper critical field $H_{c2}^{\rm orb}(0) = -0.693T_c(dH_{c2}/dT)|_{T=T_c}$.

However, our analysis shows that it strongly deviates from the experimental data if the spin effect is ignored [i.e. if Eq. (2) applies], as seen in Fig. (4) from the comparison of the fits at $H_0 \parallel a\&b$ by the dashed blue ($\alpha = 0$, $\lambda_{so} = 0$) and the solid blue ($\alpha = 2.3$, $\lambda_{so} = 0$) lines, as well as from the comparison of the fits at $H_0 \parallel c$ by the dashed green ($\alpha = 0$, $\lambda_{so} = 0$) and the solid green ($\alpha = 4.3$, $\lambda_{so} = 0$) dashed lines (with cross signs). The dashed lines are the fits by neglecting the spin paramagnetic effects. Apparently, it is the solid lines that give the best fit, indicating that the spin effect may play an important role in the temperature-dependent behavior of H_{c2}.

Since the fitted values of $\alpha = 4.3$ and $\alpha = 2.3$ for $H_0 \parallel c$ and $H_0 \parallel a\&b$, respectively, are rather large and $\lambda_{so} = 0$ could be due to weak spin-orbit scattering,[46] it suggests that the spin-paramagnetic effect is the dominant pair-breaking mechanism in FeSe, which was also observed in other Fe-based superconductors.[33,37,47–50]

With an extrapolation to lower temperatures, we have the zero-temperature upper critical field $H_{c2,\parallel c}(0) = 12$ T and $H_{c2,\parallel ab}(0) = 29$ T, at $H_0 \parallel c$ and $H_0 \parallel a\&b$, respectively. These values match those in earlier reports by other measurement techniques on it and similar materials.[9,18,51–53]

Since the Pauli-limit field is defined by $H_{\rm pauli}(0) = \Delta/(\sqrt{2}\mu_{\rm B})$,[54] where Δ is the superconducting energy gap and $\mu_{\rm B}$ is the Bohr magneton, we estimated the Pauli limit field $H_{\rm pauli}(0)$ of FeSe to be 15.9 − 30.5 T considering the previously reported value $\Delta = 1.3 - 2.5$ meV.[16,55,56] Therefore, the extrapolated values of $H_{c2,\parallel c}(0)$ and $H_{c2,\parallel ab}(0)$ in our experiment are close to and within the range of the Pauli limit, at $H_0 \parallel c$ and $H_0 \parallel ab$-plane, respectively.

Furthermore, the ab-plane and c-axis coherence lengths at $T \to 0$ [$\xi_{ab}(0)$ and $\xi_c(0)$] can also be estimated from the anisotropic Ginzburg-Landau expression $H_{c2,\parallel c}(0) = \Phi_0/[2\pi\xi_{ab}^2(0)]$, and $H_{c2,\parallel ab}(0) = \Phi_0/[2\pi\xi_{ab}(0)\xi_c(0)]$,[29] where $\Phi_0 = 2.07 \times 10^{-15}$ Wb is the flux quantum. These yield $\xi_{ab}(0) \sim 5.2$ nm and $\xi_c(0) \sim 2.2$ nm for our sample, which also agree with previous findings.[9,51,55] This gives an anisotropy parameter $\Gamma(0) = H_{c2,\parallel ab}(0)/H_{c2,\parallel c}(0) = \xi_{ab}(0)/\xi_c(0) \sim 2.4$, indicating that the electronic properties of FeSe are rather anisotropic. Nevertheless, the c-axis penetration depth $\lambda_c(0) \sim 1054$ nm can be obtained from the anisotropy relation[29] $\xi_c/\xi_{ab} = \lambda_{ab}/\lambda_c$, as the reported in-plane penetration depth $\lambda_{ab}(0) \sim 445$ nm.[18,57]

A comparison of T_c, upper critical field, anisotropy and coherence length at $T \to 0$ of our FeSe samples with those of other typical Fe-based superconductors[33,35–45] is shown in Table I. It can be found that FeSe has a higher anisotropy than many other Fe-based superconductors but with lower values of T_c and upper critical field. The coherence length of FeSe is similar to that of others.

IV. CONCLUSIONS

In summary, we measured the superconducting anisotropy of FeSe utilizing the resonance frequency of a capacitance tuning LC circuit designed by ourselves. We found that the temperature dependence of the upper critical field $H_{c2}(T)$ for both field directions at $H_0 \parallel c$ and $H_0 \parallel ab$-plane could not match the WHH model[28] when ignoring the spin effect. However, they coincided by taking into account of the Maki parameter α, suggesting an important spin-paramagnetic effect as the pair-breaking mechanism in the superconductivity of FeSe. Based on the WHH model,[28] the extrapolated zero-temperature upper critical fields $H_{c2,\parallel c}(0)$ and $H_{c2,\parallel ab}(0)$ were found to be close to and fall within the range of the Pauli limit at $H_0 \parallel c$ and $H_0 \parallel ab$-plane, respectively. The coherence length, penetration depth and the anisotropy relation were also analyzed and compared with those of other typical iron-based superconductors. A lower upper critical field and higher superconducting anisotropy at zero temperature were found in FeSe. These results match those in earlier reports by other measurement techniques on it and similar materials,[9,18,26,27,51–53] verifying that the novel resonance frequency technique is a valuable sensitive probe in detecting rich properties of unconventional superconductors.

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